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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 5 : B32B 31/00	A1	(11) International Publication Number: WO 95/13920
		(43) International Publication Date: 26 May 1995 (26.05.95)

(21) International Application Number: PCT/US93/11053	(81) Designated States: CA, US. Published <i>With international search report.</i>
(22) International Filing Date: 16 November 1993 (16.11.93)	
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(54) Title: PREPARATION OF WOOD LAMINATES

(57) Abstract

A method for making laminated wood products, particularly laminated veneer lumber, and especially from wood veneer of high moisture contents wherein the veneer is bonded together using a freely-flowing, finely divided spray-dried, powdered adhesive containing a phenolic resole resin.

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PREPARATION OF WOOD LAMINATES

FIELD OF THE INVENTION

This invention is directed to a method for preparing laminated wood products such as plywood and laminated veneer lumber (LVL). The present invention is specifically directed to preparing such laminated wood products by consolidating a plurality of wood veneers using a dry, finely powdered phenol-formaldehyde resin-based adhesive.

BACKGROUND OF THE INVENTION

Powdered adhesives, such as made from spray-dried urea-formaldehyde and phenol-formaldehyde resins, have been used widely to produce waferboard and particleboard from such wood sources as wood wafers, wood chips, wood flakes, wood shavings, wood splinters, saw dust and the like.

U.S. Patents 4,098,770 and 4,424,300, for example, describe the preparation of spray-dried phenolic resole resin-based powders for preparing adhesives for making such bonded wood products.

According to U.S. 4,098,770 the resole resin is spray dried, preferably in the presence of a non-phenolic polyhydroxy compound, to produce a free flowing powder which can be used directly or reconstituted as a liquid by adding the desired amount of water. The dry powder is said to be stable for at least six months, and can be used in place of a liquid adhesive for making waferboard and chipboard at shorter pressing times and lower pressing temperatures. Notably, when exemplified for producing plywood, the powder is first reconstituted with water to produce an aqueous adhesive which then is applied to the plywood veneer. (Example 6).

U.S. 4,424,300 proposes to make a powdered resin adhesive by spray drying a liquid mixture of novolak and resole phenol-formaldehyde prepolymers. The resin powder is disclosed for use only as a waferboard binder. Use of the powdered resin for plywood manufacture is not described or suggested.

In recent years, major gains have been achieved in reducing plywood hot press times due to the development of faster curing resins. There also have been claims of increased productivity of wood veneer driers purportedly due to the development of more moisture tolerant aqueous-based phenol-formaldehyde adhesives.

At the present time, commercial production of plywood, especially exterior grade plywood, and other laminated wood products, such as LVL, rely exclusively on the use of liquid adhesives (solutions and suspensions). Liquid compositions containing thermosetting phenol-formaldehyde resole resins are the adhesives of choice for such products, particularly for exterior grade plywood products. The present invention, in contrast, proposes to use a powdered phenolic resole resin-based adhesive in such applications. In particular, the present invention is founded on the use of a dry, freely flowing, finely divided powder adhesive based on a phenolic resole resin. This dry adhesive can be used at reduced adhesive spreads and faster press times, relative to the presently employed aqueous liquid adhesives, to produce laminated wood products, especially laminated wood products from veneers of more elevated moisture contents, while yielding bond strengths (measured as percent wood failure) on a par with laminated wood products made using liquid resole resin-based adhesives, even from veneers of lower moisture contents.

DESCRIPTION OF THE INVENTION

According to the present invention, laminated wood products are prepared by bonding together a plurality of wood veneers with a dry, freely flowing, finely divided particulate or powdered phenolic resole resin-based adhesive. Such laminated wood products include plywood and laminated veneer lumber or LVL.

Plywood is a panel-type product prepared by compressing, with an adhesive binder, large sheets of relatively thin wood veneer or plies. The normal

construction has an odd number of layers with the grain of adjacent layers at an angle to one another (most often 90°). Inner plies are commonly referred to as the core and the outer pieces as faces, front and back. The core generally is made of wood veneer, but may be lumber or even particleboard. Total panel thicknesses are usually not less than about one-eighth inch nor more than about two inches. At the present time, more than about 95% of all plywood produced is less than about one inch thick.

Using current commercial plywood manufacturing procedures, a plurality of wood veneers are coated with an aqueous adhesive and pre-assembled into multi-ply sheets. The pre-assembled sheets are held in open assembly at ambient temperature until a full press load is prepared for the hot press cure cycle. This operation may require a half hour or more. During this open assembly time, the wood absorbs water from the aqueous adhesive glue. With a highly absorbent softwood, such as the spring wood portion of Southern yellow pine, rapid dehydration of the glueline may occur with resultant inadequate glue flow and inadequate penetration during the subsequent hot press cure. With a less absorptive wood veneer, like Douglas fir veneer, an aqueous glueline dries more slowly and too much flow and penetration may occur during hot pressing. Either condition can lead to substandard plywood.

The present invention uses a dry, freely flowing, finely divided powdered adhesive which by its nature avoids the problems of premature dry-out or adhesive over penetration. As described more fully hereinafter, use of a powdered adhesive also allows more control over the molecular weight of the phenolic resin. Liquid adhesives, based on aqueous phenolic resole resins, have a limited shelf life due to the continued and inherent resin advancement during storage. Thus, during the preparation of such adhesives, one is limited with respect to the permissible molecular weight range of the resin product. Powdered resin adhesives have a broader molecular weight tolerance because of their inherent stability during storage. Molecular weight is one of the stronger influences on the flow characteristics and thus the performance of the adhesive resin during hot press curing.

LVL billets normally are constructed with the grain direction of all the veneers aligned parallel to one another. Though some LVL products are as thin as three-quarters of an inch ($\frac{3}{4}$ "), the predominant commercial constructions are one and one-half inch ($1\frac{1}{2}$ ") thick and greater. Consequently, while plywood may be prepared in 3-ply to 6-ply constructions, most LVL products generally have over 10-15 plies.

LVL billets are often produced in continuous lay-up operations and are cut to length after they exit from the hot press. Alternatively, they may be constructed in predetermined lengths. These lengths are virtually all greater than 30 feet often up to 80 feet. Inner ply veneers of LVL are often intentionally lap jointed or gap butt jointed for improved strength and to provide steam vents to reduce blows. Selected veneers may also be scarf jointed to provide adequate continuous veneer length for the billets being constructed. Plywood is virtually never produced in a continuous lay-up process and most of what is produced is laid up in 4 feet wide by 8 feet long sheets. Also, in plywood lay-up line operations, the panels are cut to length prior to the actual hot pressing operation.

As used herein, the term "veneer" refers to thin sheets of wood produced for example by skiving a thin wood layer or sheet from a log. Generally, a veneer panel or sheet will have a thickness of about 0.1 inch (2.54 mm) to about 0.167 inch (4.233 mm). Veneer is provided in 4 feet x 8 feet sheets for plywood preparation, although larger or smaller dimensions obviously can be employed. The dimension of veneer used in LVL production generally can be more variable. Representative wood varieties which can be used to prepare veneers for practicing the present invention include soft woods such as Southern pine, Douglas fir, Ponderosa pine, and the like, and hard woods such as oak, walnut and birch. Veneer for laminated wood products can be prepared by heat conditioning straight logs cut to length in vats containing hot water, and then continuously peeling the thin veneer from the heated logs. Thereafter the veneer is dried to the desired moisture content.

LVL is almost exclusively made with softwood veneer; plywood, in comparison, often is made from softwood, mixed softwood and hardwood, and

hardwood veneers. While plywood may be made using veneers of various thicknesses arranged throughout the panel, LVL tends to be prepared using a veneer of one thickness.

In accordance with current commercial plywood production procedures, wood veneers used to make wood laminates generally have moisture contents of less than about 10-15% based on the dry weight of the wood. At the present time, LVL is prepared almost exclusively from veneer dried to a total moisture content of less than about 7%. Unless there is careful control by the plywood or LVL mill operator, use of high moisture content veneers in conventional processes invariably leads to a large number of lamination defects and an excessively large number of rejected panels because of poor lamination. The lamination defects are caused by steam formation between veneer layers and a blow out of the steam when pressure is released upon completion of the pressing cycle. The water contributing to this steam formation originates both with the wood and the adhesive. As the temperature increases in the center of the veneers during consolidation, so does the vapor pressure of trapped steam. As the press is opened, the built-up vapor or steam seeks an avenue of escape and may blow the panel. Sizable operating and capital costs have been incurred in the prior art to assure a consistent moisture content for wood veneers used in making wood laminates so as to eliminate lamination defects and reduce the number of rejected panels.

Until recently, continuous commercial production of acceptable laminates from wood veneers having overall moisture contents of greater than about 5 to 10% had not been possible using conventional operating procedures. With the more recently developed faster curing liquid resin adhesives claims have been made that good bonding can be obtained using higher moisture content veneers. Even as described, however, this art typically teaches that the veneer must be dried to a total moisture content of less than about 15% and usually less than about 12%. In actual practice, use of such materials and conditions on a routine basis to obtain satisfactory bonding results has not been possible. As will be described in detail hereafter, the dry, freely-flowing, finely divided, spray-dried phenol-

formaldehyde resole resin-based adhesive of the present invention, however, makes it possible to prepare acceptable wood laminates from high moisture content veneer, i.e., a plurality of wood veneer each having an overall moisture content of greater than about 15%.

In conventional, continuous lay-up LVL operations, open assembly time prior to hot press entry generally is designed to be less than about five minutes. Often no pre-pressing operation is performed. In single billet lay-up LVL operations, as opposed to the continuous lay-up, open assembly time prior to pre-pressing is also designed to be as short as possible, typically less than five minutes. Problems frequently occur during such batch processing, however, that can stretch this time out to about 15 minutes or more. Stand time after assembly but before pre-press can range from negligible to five minutes. Pre-pressing time generally runs 1-5 minutes. When using the present invention, whether for plywood or LVL, no prepress operation is needed. As such, the open assembly time prior to hot pressing is significantly shortened and productivity is accordingly increased.

When producing a laminated wood product, such as plywood and LVL, the most critical glueline during the hot press cure operation is the innermost glueline of the core. The press conditions, temperature pressure and particularly time, must be controlled to ensure the complete cure of this glueline, since it will not be fully cured as rapidly as the other gluelines are. Not surprisingly, the press time needed to obtain full cure of all the gluelines, especially including the innermost glueline, is to a large extent a function of the panel construction. For example, plywood prepared using conventional liquid resole resin-based adhesives from 3 plies of 1/8 inch veneer may be fully cured at a press time of about 3 minutes, while 5 ply product may take 4.5 to 6.0 minutes. Use of a dry, freely flowing, finely divided powdered phenol-formaldehyde resole resin-based adhesive, however, improves the heat transfer during the press operation and thus allows the innermost glueline of laminated wood products to achieve a fully cured condition in about a 10-25% shorter time period than now practiced commercially with liquid adhesive resins.

Plywood generally is hot pressed at temperatures of 135°-180°C (about 275-360°F), most often at temperatures of about 140°-165°C (about 285-330°F), and such conditions are appropriate for the present invention. Hot press pressure is almost never greater than about 14 kgm/cm² (about 200 PSI) and, most recently, is being staged in commercial operations (high pressure to low pressure) once during the cycle to maximize panel thickness. A typical staging would be an initial pressure of about 10.5-12.3 kgm/cm² (about 150-175 PSI), reduced early in the cycle to about 7.7-9.1 kgm/cm² (about 110-130 PSI). Again, such press pressures can be used in connection with the present invention.

LVL is generally hot pressed at temperatures of about 150°-190°C (about 300-380°F), most often at temperatures of about 155°-180°C (about 315-360°F). Hot press pressure is generally varied throughout the duration of the pressing cycle. Pressures as high as about 19 kgm/cm² (about 275 PSI) for a significant part of the cycle are often used. Press pressure is controlled to maintain thickness control in operations that employ lapped veneer innerply lay-up procedures. These commercial conditions remain suitable for use in connection with the present invention. Due to the thicker constructions, press times generally are longer for LVL as compared to plywood. However, for the same reasons as discussed above, the improved heat transfer realized using the finely divided, dry powdered adhesive of the present invention permits a significant reduction (typically 10 to 25%) in the hot press time needed to produce a LVL product.

Conventional adhesives for laminated wood products comprise aqueous mixtures usually containing from about 28 to 38% active phenolic resole resin solids and 5 to 20% additional solids of various amylaceous and fibrous fillers/extenders. Powdered vegetable materials such as starch, nutshells (for example, walnut shells, pecan shells, coconut shells, ivory nut shells, horse chestnut shells, peanut shells, and the like), wood flours, barks, leaves, corn cob (Co Cob), rice hull and the like are usually mixed into the adhesive as fillers either alone or in combination with a dilutent such as wheat flour (Glu X), sorghum flour and the like. Such additives are used for reducing the adhesive cost, for preventing over penetration of liquid adhesive into the wood veneer, for retaining

a uniform liquid adhesive viscosity to facilitate the spreading of the adhesive on the veneer surface, and for preventing the formation of interstices and cracks accompanying the shrinkage and aging of the cured resin after adhesion. Fillers often are used in an amount of from about 8 to 14% based on the weight of the phenolic resin. Inorganic extenders such as attapulgite clay also can be used. Although not needed for many of the same reasons, such additives can also be used in connection with the present invention primarily to reduce adhesive cost.

Depending on the wood species, prior art aqueous adhesive application rates often ranged between about 160 and 270 grams per square meter (0.033 to 0.055 pound per square foot) of single glue line. This corresponds to an application level based only on resin solids of typically between about 45 to 103 grams per square meter (0.009-0.021 pound per square foot) of single glue line. Another advantage of the present invention is that laminated wood products of a comparable strength can be prepared using a glue spread, based on resin solids, of as much as 15 to 45% lower than when using a liquid adhesive. Glue spreads, based on resin solids, useful in the present invention generally will vary between 30 and 90 grams per square meter.

Aqueous phenol-formaldehyde resins useful for preparing the solid dry, freely-flowing, finely divided particulate or powdered resole-based adhesives of the present invention constitute thermosetting condensation products produced by reacting at least an equal molar amount of formaldehyde with phenol. Preferred phenol-formaldehyde condensation products are prepared using a molar ratio of phenol to aldehyde in the range of about 1:1.5 to 1:3 with a phenol to formaldehyde molar ratio of about 1:2 usually being preferred. Suitable aqueous resole resins are prepared by reacting phenol and formaldehyde at these molar ratios in an aqueous reaction medium in the presence of an alkaline catalyst. In the broad practice of the present invention, standard reaction conditions, procedures and reactants widely used for preparing aqueous phenol-formaldehyde resole resins can be used.

Sodium hydroxide generally is the alkaline catalyst of choice in the present invention. A list of other potential alkaline catalysts includes, *inter alia*, other

alkali metal hydroxides such as potassium hydroxide, alkali metal carbonates such as sodium and potassium carbonates, and alkaline earth metal oxides and hydroxides such as barium hydroxide, calcium hydroxide and calcium oxide.

A variety of techniques are known in the art for reacting phenol and formaldehyde in the presence of the alkaline catalyst. Typically, the resin is reacted in stages with separate partial additions of either one or both of the reactants and the alkaline catalyst. For example, one common procedure is to react the phenol with a portion of the formaldehyde, in the presence of a portion of the alkaline catalyst. After a brief, initial exothermic reaction, additional amounts of alkaline catalyst and formaldehyde are added to the reacting mixture and the reaction is continued with careful control of the reaction temperature. Once all of the reactants and catalyst have been added, the reaction is allowed to proceed to a suitable end point, which may be determined by measuring the refractive index of the reacting mixture or by measuring the viscosity of the reacting mixture or by some combination thereof as recognized by those skilled in the art. Once the selected end point is achieved, the reaction mixture is ready for preparing a dry, freely-flowing, finely divided powdered adhesive mixture by spray drying.

The amount of alkaline catalyst added during resin preparation to produce a resole resin solution is often sufficient to produce an alkalinity content, in the range of about 1% to about 15%, usually about 3% to about 9%, and most often about 4% to about 8% based on the weight of the resole resin solution. As used herein alkalinity content means the content of alkaline catalyst in the resin solution expressed as a weight percent of the solution on the basis of the equivalent (mole) weight of sodium hydroxide. For example, in the case where the aqueous resin solution contains about 6.4 % by weight potassium hydroxide, the alkalinity content of the resin solution would be about 4.6% based on the equivalent (mole) weight of sodium hydroxide since potassium hydroxide has a greater molecular weight than sodium hydroxide. Normally, from about 50 mol % of sodium hydroxide up to about 100 mol % of sodium hydroxide based on the mols of phenol used in preparing the resole resin will provide a desired level of alkalinity.

Usually about 0.60 mol to about 0.80 mol of sodium hydroxide per mol of phenol is used.

Preferably, hydroxybenzene (phenol) is the phenol reactant of choice for making the aqueous resole resin, although substituted phenols such as cresol and higher functional phenols such as resorcinol or bisphenol-A, can be used. Formaldehyde is the preferred aldehyde constituent. Generally, the formaldehyde is supplied as an aqueous solution known in the art as "formalin". Formalin is an aqueous solution that generally contains from about 37% to about 50% by weight formaldehyde. Other forms of formaldehyde such as paraformaldehyde also can be used. Other aldehydes, which may be used in combination with formaldehyde, include aliphatic aldehydes such as acetaldehyde and propionaldehyde; aromatic aldehydes such as benzylaldehyde and furfural and other aldehydes such as aldol, glyoxal and crotonaldehyde.

One advantage of the present invention is that the molecular weight of a resole resin suitable for preparing the dry, freely-flowing, finely divided particulate or powdered resole resin-based adhesive by spray drying in accordance with the present invention can be higher than for a liquid resole resin that is to be stored for subsequent use in formulating an aqueous adhesive. This is because the spray-dried, heat hardenable resin solids are more stable than the precursor aqueous thermosetting resin, which generally has a limited shelf life due to the inherent molecular weight advancement that occurs on storage of such aqueous resins. Thus, when used in connection with the present invention, the aqueous resole resin prior to spray drying can normally be advanced to a higher molecular weight end point than is conventional for aqueous resole resins prepared for plywood and LVL adhesive applications. The desired molecular weight of the spray-dried resin solids will be influenced to a large extent by the veneer moisture content. At higher veneer moisture contents, a resin with a higher molecular weight and lower flow characteristics often will be favored.

For preparation of laminated wood products according to the present invention, aqueous resole resins, prior to spray drying, may exhibit a number average molecular weight in the range of about 2,000 to 10,000 and a weight

average molecular weight of about 20,000 to 50,000. The aqueous-based resin's polydispersity often is between about 7 and 14. For purpose of the present invention, the weight and number average molecular weights of the phenol-formaldehyde resole resin can be determined by gel permeation chromatography (GPC) using tetrahydrofuran (THF) solvent, polystyrene standards and procedures well known to those skilled in the art.

The process of making the spray-dried precursor, aqueous resole resin of the present invention is directly adaptable to equipment conventionally used for making phenol-formaldehyde resole resins. As noted above, the reaction is conducted in aqueous solution. Normally, the reaction is conducted so that the ultimate resole resin has a non-volatile material (NVM) content of at least about 35% by weight based on the weight of the resole resin solution. NVM contents of up to about 55-60% are possible, although the NVM content often does not exceed about 50% in order to avoid solutions that have such high viscosities that they present problems in mixing, pumping and spray drying.

In using the resole resin for preparing a dry, freely-flowing, finely divided particulate or powdered, spray-dried resole resin-based adhesive useful for bonding a laminated wood product such as plywood or LVL, it is common to blend the aqueous resin with additional ingredients before spray drying. In addition to the resin solution itself, which constitutes the major component of the aqueous adhesive mixture (generally from about 75 to 100% by weight of the adhesive solids), it is common to include the various fillers, i.e. inert materials added to increase the weight of the adhesive mixture, adhesive extenders, additional caustic and other known adjuvants as indicated to a certain hereinabove. Generally, a suitable aqueous adhesive before spray drying will contain from about 25 to 50% resin solids, from about 3 to 7% fillers, from about 3 to 7% extenders and from about 1 to 10% additional alkaline catalysts. As noted above, suitable fillers and extenders include starch, wood, flour, nut shell flour, bark products or agricultural residues, clays, and corn husks. Starch and clays generally are used as fillers, often in amounts of 1% to 10% based on the weight of the resin solids. On a water-free basis the adhesive generally contains about 75 to 100% resin solids.

Resin catalysts for accelerating the curing (hardening) rate of the resole resin, such as resorcinol-formaldehyde resins and phenol-resorcinol-formaldehyde resins, also can be added. Such ingredients may be added to the aqueous adhesive before spray drying or they can be added as spray dried powders themselves by dry blending subsequent to the spray drying of the aqueous phenolic resole resin. These resins are known catalysts for accelerating the cure rate of phenol-formaldehyde resole resins. The preparation and use of these resins in the context of formulating adhesives for making wood laminates is well-known to those skilled in this art.

The aqueous resol resin-based adhesive then is fed to a spray drier for producing the dry, freely-flowing, finely divided particulate or powdered adhesive used in accordance with the present invention. As used herein, the term "spray dryer" refers to the technically sophisticated process of atomizing (in the form of finely divided droplets) a solution or slurry into a hot gas stream under controlled temperature conditions and under specific gas/liquid contacting conditions to effect evaporation of water from the atomized droplets and production of a dry freely flowing, finely divided particulate product.

Spray drying, as used herein, is typically carried out with pressure nozzles or centrifugal atomizers operating at speeds of up to 10,000 to 16,000 RPM or more. At these speeds, one milliliter of liquid feed can be converted to over 100 million fine droplets. Despite this high velocity generation of droplets, the spray dryer is designed so that the droplets do not contact the spray dryer wall under proper operating procedures. This effect is achieved by a precise balance of atomizer velocity, air flow, spray dryer dimensions of height and diameter, and inlet and outlet means to produce a cyclonic flow of air in the chamber. This highly specialized art is clearly to be distinguished from the random and promiscuous spraying of liquid droplets such as one might do with a garden hose.

In spray drying, the liquid adhesive feed is converted to a fine spray; the water in the feed is evaporated by means of a stream of hot gas, usually air; and the dry, powdered product is separated from the stream of hot air. Moisture evaporation is controlled by controlling of the inlet and outlet temperature of the

hot air used for drying. Due to the thermosetting nature of the product, it is preferred that the outlet temperature be regulated within the range from about 70° to 90°C. Variables such as droplet size, air flow rate, air temperature and humidity also affect the rate of drying and ultimate moisture content of the powdered product. The particle size distribution of the spray-dried adhesive powder, which varies with the degree of atomization of the liquid adhesive and amount of solids in the liquid feed, should preferably be from about 20 microns and up, with no particles larger than about 60 microns. A spinning disc-type atomizer is preferably used for spray drying the liquid adhesive. Before storing the particulate adhesive, it is desirable to cool the material quickly to room temperature. This can be accomplished by contacting the powdered adhesive with chilled air.

The freely-flowing, finely divided resole resin-based adhesive powder of the present invention is to be distinguished from dry, pulverized phenolic condensation product referenced in U.S. 2,150,697. When used in connection with dried heat-hardenable resole resins, pulverization procedures likely cannot provide the degree of subdivision without undesired resin advancement realized via spray drying and essential to the successful practice of the present invention. Moreover, the use of such pulverized material even for the preparation of plywood, with a small number of veneer plies, is far afield from the preparation of laminated veneer lumber with 10 or more plies using a spray-dried adhesive.

Laminated wood products can be prepared from the spray dried phenol-formaldehyde resole resin-based adhesive by applying a thin, substantially uniform layer of the freely flowing particulate dry adhesive to the mating surfaces of a plurality of wood veneers. The powdered adhesive can be applied using any one of a wide variety of solids handling means for laying down a uniform layer of the powdered adhesive material onto the mating surface of each veneer. Such equipment would include devices that operate on gas entrainment such as pneumatic spray guns or curtain-type coaters with spaced nozzles or orifices and devices that operate with mechanical/gravity type powder feed such as a sifting

means. Leveling means may be included to ensure that a substantially uniform layer of adhesive is applied to each veneer mating surface.

Depending on whether plywood or LVL is being prepared the veneer sheets with the applied layer of adhesive are assembled into a laminate structure so that the powdered adhesive is sandwiched between mating surfaces of the wood veneers. The laminate then is consolidated at a temperature and pressure and for a time sufficient to cure the resole resin, as described above in connection with plywood and LVL manufacturing procedures.

A particularly surprising feature of the present invention is that a plurality of wood veneers each with total moisture content of 15% and above can be successfully bonded into a laminated wood product using an adhesive application level of 30 to 90 grams of resole resin solids per square meter of single glue line.

The following examples are presented to illustrate and explain the invention. Unless otherwise indicated, all references to parts and percentages are based on weight and all temperatures are expressed in degrees Celsius.

Example 1

An aqueous resole resin suitable for spray drying to produce a freely-flowing, finely divided powdered adhesive can be prepared as follows. About 25 parts phenol is added to a suitable reaction vessel. Then about 13 parts of a 50% aqueous formaldehyde solution, 26 parts additional water, 0.3 parts cresylic acid, a minor amount of a defoaming agent, 0.4 parts pearl starch and about 5.7 parts of 50% solution of sodium hydroxide (caustic) are added. The reaction mixture is allowed to exotherm to about 80°C and the reaction mixture is held at that temperature for about 10 to 20 minutes. Additional formaldehyde is then slowly added over about a 3/4 to 1 hour period while maintaining the batch at a temperature of about 80°C to increase the formaldehyde to phenol (plus cresylic acid) mole ratio to about 2.2:1. The batch is then heated quickly to about 95-100°C and held at that temperature until the reaction mixture exhibits a Gardner-Holt (G-H) viscosity of about A-B. The batch is thereafter cooled to about 80°C and held until a G-H viscosity of about R-S is reached. About 5.7 parts additional caustic is added and the batch is reacted to a G-H viscosity of about U-V. Once

the target viscosity is reached, the batch is cooled and can be stored for a short time before spray drying. The resin solution typically contains about 43% solids, has an alkalinity content of 5.7 and a Brookfield viscosity of about 1200 cps.

Example 2

LVL billets (15 ply - 1.5" thick) were produced by applying a powdered phenolic-formaldehyde resole resin prepared by spray drying an aqueous resole resin to the veneer in an amount of about 43 grams per square meter of single glueline. The billets were hot pressed using typical LVL press conditions to cure the resin. Wood failure rates, using American Plywood Association (APA) test procedures for LVL, were 93 to 98%.

Example 3

Douglas Fir veneer, one-tenth inch thick, was used to prepare 1½ inch-15 ply panels with the dimensions 12 inches by 12 inches. GP 5478 phenolic resole resin, a spray dried aqueous resole waferboard resin, commercially available from Georgia-Pacific Resins, Inc., was applied to the veneer using a salt shaker-type applicator. Four panels were fabricated at a resin application level of about 43 grams per square meter (4 gm per ft²) and four panels were fabricated at a resin application level of about 64 grams per square meter (6 gm per ft²). Another four panels were made by applying the resin at 43 grams per square meter (4 gm per ft²) followed by a water spray on the glue line. Finally, a final set of four panels were prepared by applying GP 52C59 resin spray dried aqueous resole waferboard resin, commercially available from Georgia-Pacific Resins, Inc., at an application level of about 43 grams per square meter (4 gm per ft²). The following table summarizes other processing parameters. Based on a visual examination of the bond quality, the majority of the panels had a good wood failure in the core.

Table 1.
LVL Panel Properties

Resin	Panel #	Wood Failure %		Resin used (gms/ft ²)	Time to 220°F (minutes)	Max. temp. °F
		Top	Cntr.			
GP 5478 @ 4 gms/ft ²	429	99	100	4.3	16.4	240
	430	92	92	4.4	15.7	246
	431	100	95	4.5	16.1	242
	432	100	100	3.7	17.0	234
	AVG	98	97			
GP 5478 @ 6 gms/ft ²	433	52	13	6.1	15.4	249
	434	80	11	6.4	15.1	251
	435	80	58	6.2	15.1	239
	436	94	74	6.5	15.0	252
	AVG	77	39			
GP 5478 @ 4 gms/ft ² with water spray	453	93	100	4.7	14.7	249
	454	98	100	4.1	16.0	237
	455	98	92	4.2	14.9	249
	456	90	99	4.3	14.3	250
	AVG	95	98			
GP 52C59 @ 4 gms/ft ²	449	96	100	4.9	15.1	255
	450	100	99	3.9	16.7	249
	451	92	98	4.4	14.4	257
	452	88	98	4.7	14.8	244
	AVG	94	99			

Example 4

An aqueous resole resin suitable for spray drying to produce a freely-flowing, finely divided powdered adhesive can be prepared as follows. Phenol (about 29 parts), formaldehyde (about 10 parts of a 50% aqueous solution) and about 23 parts water are added to a suitable reaction vessel. Then about 6.3 parts of a 50% aqueous sodium hydroxide solution (caustic) is added and the reaction mixture is heated, at least in part by the reaction exotherm, to a temperature of about 85°C. Additional formaldehyde is added in an amount sufficient to bring the formaldehyde to phenol mole ratio to about 2.2:1 and the batch is reacted to a G-H viscosity of about B. The batch then is cooled to a temperature of about 80°C, reacted to a G-H viscosity of EF, cooled quickly to a temperature of about 65°C and reacted to a G-H viscosity of about J. A small amount of tall oil fatty acid (TOFA) and ammonium hydroxide (1 part of a 28% aqueous solution) are added during the cooling step. As produced, the resin solution has a solids content of about 45%, an alkalinity content of about 3.5% and a Brookfield viscosity of about 275 cps. The resin is suitable for spraying to a freely-flowing, finely divided powder.

While certain specific embodiments of the invention have been described with particularity herein, it will be recognized that various modifications thereof will occur to those skilled in the art and it is to be understood that such modifications and variations are to be included within the preview of this application and the spirit and scope of the appended claims.

CLAIMS

We claim:

1. A method for producing a wood laminate from a plurality of wood veneers comprising (a) applying a freely-flowing, finely divided particulate dry adhesive to at least one mating surface of each of said plurality of wood veneers, said adhesive having been prepared by spray drying an aqueous adhesive mixture containing a phenol-formaldehyde resole resin, said resin having been prepared as an aqueous resole resin by reacting at least an equal molar amount of formaldehyde with phenol in the presence of an alkaline catalyst, said freely-flowing, finely divided particulate dry adhesive being applied to said mating surfaces in an amount to provide 30 to 90 grams of resole resin solids per square meter of single glue line, (b) assembling said plurality of wood veneers into a laminate so that said particulate dry adhesive is sandwiched between mating surfaces of said wood veneers and (c) consolidating said laminate at a temperature, pressure and for a time sufficient to cure the resole resin.
2. The method of claim 1 wherein each of said wood veneers have a total moisture content of above about 15%.
3. The method of claim 2 wherein said alkaline catalyst is present in said aqueous resole resin in an amount sufficient to provide a resole alkalinity content of between 1 and 15%
4. The method of claim 1 wherein said freely-flowing, finely divided particulate dry adhesive has a particle size distribution with no particle larger than about 60 microns.
5. The method of claim 1 wherein said wood laminate is laminate veneer lumber.
6. The method of claim 5 wherein said laminate veneer lumber has at least 10 plies of veneer.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US93/11053

A. CLASSIFICATION OF SUBJECT MATTER

IPC(5) :B32B 31/00

US CL :156/283, 335

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 156/283, 335; 264/13; 528/147, 501; 525/480; 34/28

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

None

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

None

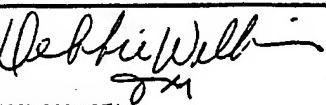
C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	GB, A, 402,677 (Thomson-Houston Co.) 07 December 1933 See Page 1 lines 15-21, 39-44 and 66-86	1-6
Y	US, A 3,342,776 (Lambuth) 19 September 1967 See col. 1 lines 12-21 and 30-35, col. 2 lines 50-63, col. 5 line 42 thru col. 6 line 25, col. 9 line 71 thru col. 12 line 50	1-6
Y	US, A, 4,098,770 (Berchem et al) 04 July 1978 See col. 1 lines 5-7 and 32-35, col. 2 line 4 thru col. 3 line 35	1-6

 Further documents are listed in the continuation of Box C. See patent family annex.

* Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be part of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document published on or after the international filing date	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubt on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means		
"P" document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search	Date of mailing of the international search report
11 JANUARY 1994	25 FEB 1994

Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	Authorized officer J. J. Gallagher  Telephone No. (703) 308-1971
Faximile No. Not Applicable	